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DETERMINATION OF NEUTRON MULTIPLICATION OF SUB-CRITICAL HEU SYSTEMS USING DELAYED NEUTRONS

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1. Introduction

The problem of determining the neutron multiplication of sub critical systems of highly enriched uranium (HEU) has always been more difficult than the equivalent problem in plutonium systems. In the latter, the driving neutron source is usually neutrons from the spontaneous fission of ^{240}Pu. The ^{240}Pu is evenly distributed throughout the volume of the plutonium. Point kinetic models[1,2,3,4] using multiplicity analysis of the observed neutron detection time distributions have been successfully applied to determine the multiplication of sub critical plutonium systems.

No comparable passive neutron technique exists for extracting attributes of HEU samples since the half-life for spontaneous fission of ^{235}U is approximately 2.5×10^{17} years. The conventional passive gamma-ray techniques, which use the 186-keV and 143-keV gamma-rays, are sensitive to only the surface of the material, and if shielding is present these gamma-rays are often unobservable. Traditionally, information about the multiplication of an HEU system has been obtained by the use of an external neutron source to induce fission in the HEU. The best situation occurs when it is possible to place the neutron source centered within the volume of the HEU. For this case the ratio of the neutron counting rates in a detector external to the HEU volume, with and without the HEU in place, determines the leakage multiplication. The determination of leakage multiplication in this manner has been shown [5] to be dependent upon the energy spectrum of the neutron source. If the center of the assembly is not accessible, the source is placed at a fixed external position opposite the neutron detector. The leakage multiplication determined in this manner is dependent upon the geometrical relationships between the HEU system, the neutron source and the detector, as well as upon the neutron source energy distribution.

The method presented here uses delayed neutrons from fission products distributed throughout the HEU sample as a source of interrogating neutrons. The fission products are from fission events produced by an external radiation probe of pulsed 14-MeV neutrons. Neutrons are detected between pulses of the interrogating probe by a medium efficiency neutron detector system. The neutron detection times are recorded, and subsequently analyzed with the Feynman reduced variance method [2,5]. This analysis provides a measure of the number of "single", "double", and "triple" neutron events detected from fission events. Ratios of doubles/singles and triples/singles are compared to model calculations to extract values for the multiplication of the HEU sample. The use of delayed neutrons to interrogate highly enric[illegible]uranium systems was first investigated by Brunson and Coop [6] using photo-fission from a [illegible]-MeV bremsstrahlung photon source as the external radiation probe.

2. Experimental Apparatus

A. External Interrogation Probe

The external radiation probe consisted of 14-MeV neutrons from a pulsed neutron generator. The neutron generator produced neutrons by means of the deuteron + triton nuclear reaction which yields ~14-MeV neutrons and ~3.4-MeV alpha particles. The neutron generator produced pulses of neutrons of approximately 20 μs width at a frequency 50 Hz. The neutron intensity was $\sim 1*10^6$ neutrons per pulse providing a total neutron output of approximately $5*10^7$ neutrons/sec into 4π steradian.

B. Neutron Detector System

The neutron detector system contained forty-eight ^{3}He gas ionization tubes, arranged into sixteen modules, each module containing three tubes. The three tubes were within a polyethylene cavity that thermalized the neutrons. The neutron detection occurs by the neutron + ^{3}He ⇒ proton +triton reaction. The ^{3}He gas pressure was two atmospheres. The half-life for neutron die away in the detector was determined to be 54.1 microseconds from a Rossi-α type time distribution measurement of a ^{252}Cf neutron source. The detector modules formed an enclosure 40 cm x 40 cm x 116 cm. The ends of the enclosure were open. The experimental geometry is shown schematically in Fig. 1. The total efficiency for detection of fission neutrons

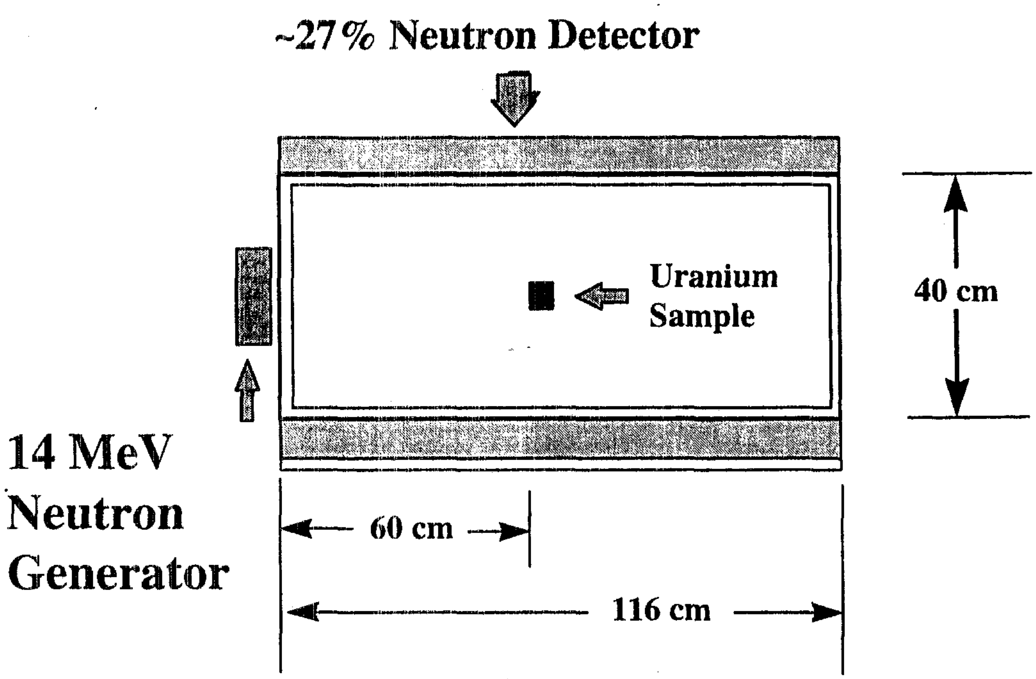


Fig. 1. Plan view of the experimental apparatus.

was approximately 27% determined from a multiplicity analysis for a ^{252}Cf neutron source located at the center of the enclosure. The HEU samples were placed within a cadmium lined box to prevent low energy neutrons resulting from room scatter of the 14-MeV neutron pulse, as well as neutrons reflected from the polyethylene boxes of the neutron detector assembly from reaching the sample. The neutron generator was located at one of the open ends of the detector 60 cm from the center of the HEU sample

Signals from the ^{3}He neutron detectors were amplified and discriminated to form logic pulses in the Los Alamos designed preamplifier, amplifier, discriminator, ECL logic pulse module, (PADEM) [7] then configured as input to the 15 channels of the custom Los Alamos designed pulse arrival time recording module (PATRM) [8]. Segmenting the detector signal reduced the effective dead time of the neutron detector from approximately 5 microseconds for a single channel to less than 0.4 microseconds. The PATRM uses a logic pulse designated VETO to control data acquisition in active mode. At the onset of the VETO pulse, the PATRM disables all data inputs, writes a code marker to its internal memory at the next two memory locations, stops the internal clock counter and resets this counter to zero. Upon lifting of the VETO, the clock counter and all data inputs are enabled. The result of the measurement is a history of the signal arrival times relative to the end of the VETO pulse. The clock frequency of the PATRM was 5 megahertz, allowing the signal arrival time to be measured with an accuracy of 0.2 μs. Up to a million events can be recorded before transferring the data to a small computer for analysis and archiving. The VETO input to the PATRM was the logic pulse of length 700 us derived from the trigger pulse from the neutron generator. The PATRM was disabled during the beam pulse and for most of the time in which the ^{3}He detectors were able to detect neutrons from the interrogating probe burst. The majority of pulses recorded by the PATRM were from delayed neutrons from fission products and neutrons from fission events induced by the delayed neutrons.

3. Highly Enriched Uranium Metal Samples

The uranium metal samples, enriched to 93.12% ^{235}U, consisted of a set of nesting hollow hemispheres of average density 18.675g/cm^3 [9]. Except for the innermost sphere which was solid with a radius of 2 cm, the wall thickness for each hemisphere was ~1/3 cm. Shells were combined to provide twelve configurations with radii ranging from 2.00 cm to 5.67 cm. The mass range was from 592 to 13722 gm. The relevant physical characteristics of the twelve configurations are listed in Table I.

TABLE I. Physical characteristics of the HEU metal shells.

Configuration #	**Radius (cm)**	**Mass (gm)**
1	2.00	592.0
2	2.34	932.7
3	2.67	1382.6
4	3.00	1961.9
5	3.34	2683.4
6	3.67	3570.8
7	4.00	4625.4
8	4.34	5868.3
9	4.67	7327.2
10	5.00	9020.0
11	5.34	10920.0
12	5.67	13722.0

4. Measurement Protocol.

Each configuration of the HEU metal shells was investigated with delayed neutrons from fission events initiated by 54,000 14-MeV neutron pulses. A small plastic scintillator viewed by a fast photo multiplier tube was placed at a distance of one meter from the neutron source and was used as a relative 14-MeV neutron flux monitor.

5. Experimental Observables, Feynman Variance Analysis

We have applied the reduced variance method of analysis developed initially by Feynman, deHoffmann, and Serber [4], and used by other investigators [1,2] of spontaneous fissioning systems. The time history of the neutron events recorded in the PATRM module, is examined to construct the Feynman histograms, FN_{ORDER}, formed by randomly opening an inspection time interval of fixed width and then counting the number of neutron signals that are present within this time interval. A histogram array is then incremented according to the number of neutrons found, and the process is repeated for the duration of the measurement. The resulting distribution then contains the number of times that zero, one, two, three, etc., neutrons were found within the inspection interval. The FN_{ORDER} distributions are converted to probability distributions P_{ORDER} by dividing the FN_{ORDER} by the number of times the inspection interval (NUM_INT) was opened.

$$P_{ORDER} = FN_{ORDER} /(\text{NUM\_INT}).$$

The first, second and third reduced factorial moments of the P_{ORDER} distributions are then formed.

$$\text{Mom1} = \Sigma\{ m*P_m \} \text{ and}$$

$$\text{Mom2} = \Sigma 1/2*\{ m*(m-1)*P_m \}.$$

$$\text{Mom3} = \Sigma 1/6*\{ m*(m-1)*(m-2)*P_m \}.$$

The first moment is the average, or mean, number of neutrons found in the time interval
The first moment divided by the time interval value (Ω) is the average neutron counting rate during the measurement,

$$\text{N1/sec} = \text{Mom1} /\Omega.$$

The second moment represents the variance of the distribution. The difference between the variance and the square of the mean is a measure of the amount of correlation present in the data. This difference divided by the inspection interval width is then the number of correlated neutrons per second, N2/sec, present in the data;

$$\text{N2/sec} = \{\Sigma\, m(m-1) *P_m - ½*(\Sigma\, m*P_m) *(\Sigma\, m*P_m)\}/ \Omega.$$

The following combination of the first, second and third reduced moments divided by the interval width is the number of triplet neutrons per second, N3/sec, present in the data,

$$N3/sec = (Mom3 - Mom2*Mom1+(1/3)*Mom1^3)/\Omega.$$

For a neutron source that has no correlation's, such as neutrons from an Americium-Lithium source, the above distributions are those of a pure Poisson distribution, and N2/sec and N3/sec are identically zero. Thus the observation of nonzero values for N2/sec and N3/sec are positive indicators of correlations in the neutron signal, which in turn is a positive indicator of neutrons from fission events.

For a passive interrogation of a system containing a spontaneously fissioning system such as ^{240}Pu or ^{252}Cf, the fission rate is essentially constant with time during the short time span of the measurement, although the time between any two fission events is completely random. For a system such as ^{235}U undergoing active interrogation, the fission rate is not necessarily constant. In order to at least partially compensate for any time dependence, we form many separate Feynman distributions. The first distribution is for an interval starting 700 μs after the neutron generator burst. The inspection window is opened once for each probe burst to form this first Feynman histogram. The inspection time interval was chosen to be 200 μs as a result of a study of the statistical precision of N2/sec and N3/sec as a function of the inspection time interval width. The second Feynman histogram begins one inspection time interval later, the third Feynman histogram two time intervals later, etc. For the data described here, the neutron generator produced a beam burst every 20,000 μs. Thus, ninety-six Feynman distributions were generated. From these Feynman distributions ninety-six values for N1/sec, N2/sec and N3/sec were calculated. Each of these distributions is for inspection intervals that are random in time with respect to any fission event.

6. Experimental Results

A. The time dependence of N1/sec, N2/sec and N3/sec

The N1/sec, N2/sec and N3/sec distributions for the 13.72 kg HEU metal sphere are shown on a log scale in Fig. 2. The data for the "singles" measurement N1/sec is shown as diamonds.

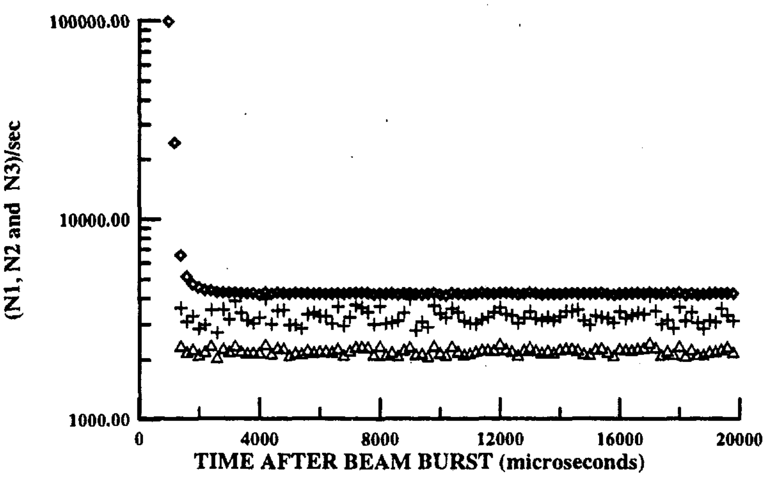


Fig 2. The time dependence of N1/sec(diamonds), N2/sec (triangles) and N3/sec (crosses) after the 14-MeV neutron beam bursts for the configuration containing 10.92 kilograms of HEU.

the "doubles", N2/sec, distribution as triangles and the "triples", N3/sec, distribution as crosses. No data is recorded during the burst and for the following ~ 700 µsecs. At the earliest times, there are a substantial number of neutrons from the neutron beam burst and from fission events induced during the burst. From 700 to ~2000 µsecs, a sharp decrease in the N1/sec distribution is observed, reflecting the decay time constant associated with the neutron detector. The results for the "background" measurement, which is with no sample in the irradiation position, are shown in Fig. 3. A sharp decrease is again observed for the N1/sec distribution from ~700 to

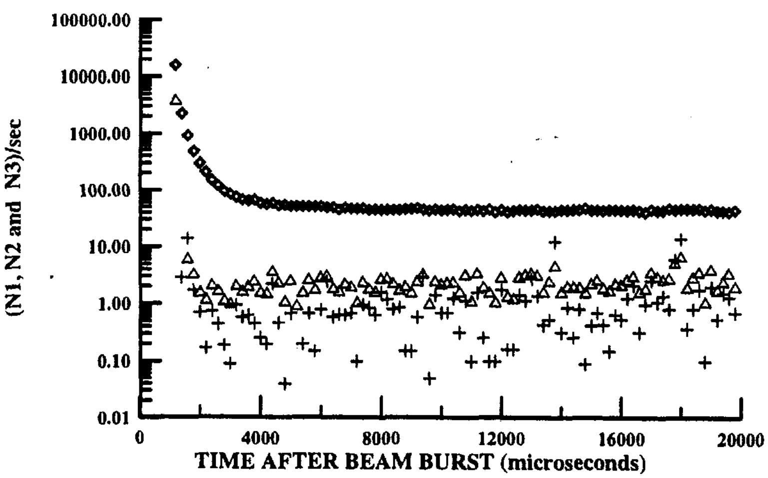


Fig 3. The time dependence of N1/sec(diamonds), N2/sec (triangles) and N3/sec (crosses) after the 14-MeV neutron beam bursts for the active background measurement with the neutron detector empty.

~3000 µsecs, and remains constant to 20000 µsecs. The net distributions, that is the background corrected distributions are shown in Fig. 4. N1/sec, N2/sec and N3/sec

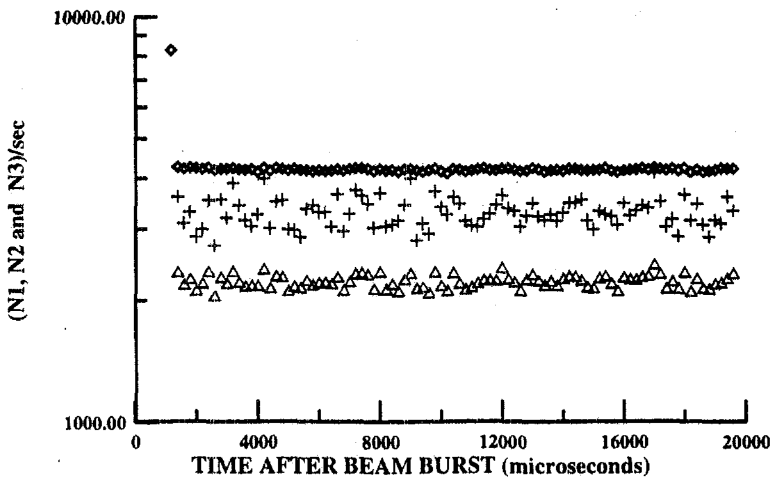


Fig 4. The time dependence of N1/sec(diamonds), N2/sec (triangles) and N3/sec (crosses) after the 14-MeV neutron beam bursts for the configuration containing 10.92 kilograms of HEU, with the active background subtracted.

are essentially constant throughout the full time of the measurement, except for the earliest data points.

Similar neutron distributions were obtained for each of the HEU metal configurations listed in Table I. After background corrections, average values for N1/sec, N2/sec and N3/sec were calculated from the data between 8000 and 16000 μsecs, and a statistical error assigned by calculating the variances about the mean for the forty independent measurements.

C. N1/sec, N2/sec and N3/sec Observables for HEU Metal Configurations

The variations of N1/sec, N2/sec and N3/sec as a function of radius are shown in Fig. 5.

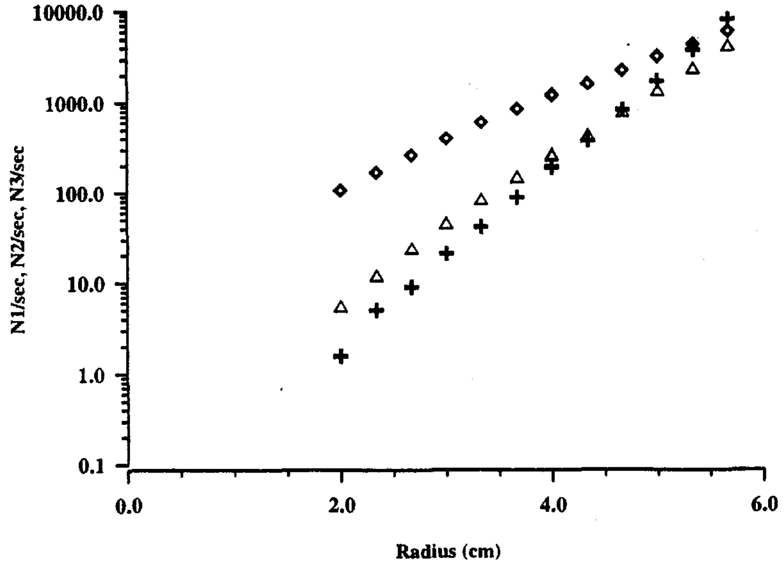


Fig. 5. N1/sec (diamonds), N2/sec (triangles) and N3/sec (crosses) as a function of radius of the HEU metal spheres.

All three observables increase with increasing wall thickness with an exponential dependence, with N2/sec increasing faster than N1/sec, while N3/sec has the fastest increase. The variation of N1/sec as a function of the HEU mass is shown in Fig. 6. A monotonic non-linear increase is

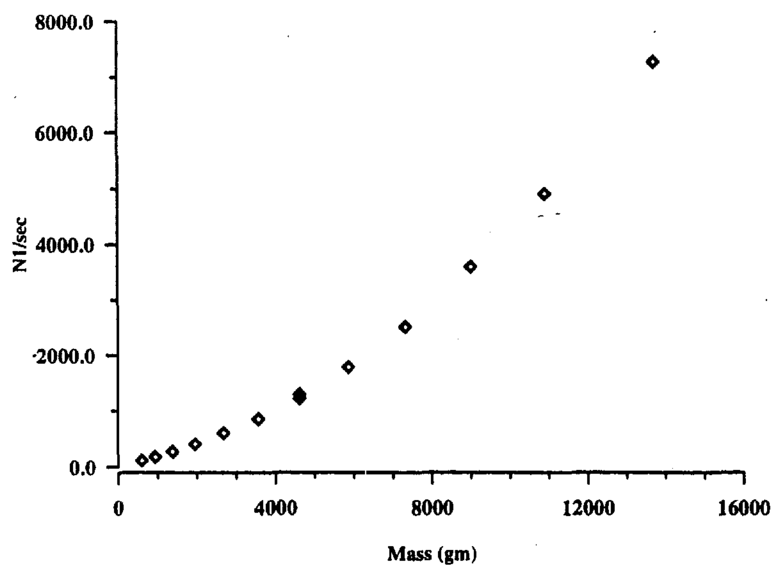


Fig. 6. N1/sec values vs. total mass of the twelve HEU metal configurations.

observed.

7. Hage-Cifarelli Model.

According to the results of Hage and Cifarelli [1] for the analysis of the spontaneous fission of ^{240}Pu mixed with ^{239}Pu, the singles doubles and triples counting rates are related to the mass, neutron detection probability, leakage multiplication, and random neutron rate via the following relations:

R1= singles counting rate,

R2 = doubles counting rate,

R3 = triples counting rate,

$R1 = \varepsilon * [\, M_L * \nu_{S1} * Fs + M_L * S\alpha\,]$,

$$R2 = (\varepsilon^2) * [\, M_L{}^2 * \{\nu_{S2} + (M_L - 1) * \nu_{S1} * \nu_{I2} / (\nu_{I1} - 1)\} * Fs + M_L{}^2 * \{(M_L - 1) * \nu_{I2} / (\nu_{I1} - 1)\} * S\alpha\,],$$

$$R3 = (\varepsilon^3) * [\, M_L{}^3 * \{\nu_{S3} + (M_L - 1) / (\nu_{I1} - 1) * [\nu_{S1} * \nu_{I3} + 2 * \nu_{S2} * \nu_{I2} + 2 * (M_L - 1) / (\nu_{I1} - 1) * \nu_{S1} / \nu_{I2}{}^2]\} * Fs + M_L{}^3 * \{(M_L - 1) / (\nu_{I1} - 1) * [\nu_{I3} + 2 * (M_L - 1) * \nu_{I2}{}^2 / (\nu_{I1} - 1)\} * S\alpha\,].$$

The parameters R1, R2 and R3 are related to the experimental Feynman observables N1/sec, N2/sec, and N3/sec by

$R1 = (N1/sec)/\Omega$,

$R2 = (N2/sec) / (\Omega * WW2)$,

$R3 = (N3/sec) / (\Omega * WW3)$, and

$WW2 = 1 - \{1 - \exp(-\lambda * \Omega)\} / \{\lambda * \Omega\}$,

$WW3 = 1 - \{3 - 4 * \exp(-\lambda * \Omega) + \exp(-2\lambda * \Omega)\} / \{2\lambda * \Omega\}$,

where
ε = fission neutron detection probability,
M_L = fission leakage multiplication,
Fs = fissions per second per gram,
$S\alpha$ = random neutron rate from alpha neutron reactions,
Tm = measurement time,
ν_{S1} = reduced first factorial moment of the spontaneous fission neutron probability distribution,
ν_{I1} = reduced first factorial moment of the induced fission neutron probability distribution,
ν_{S2} = reduced second factorial moment of the spontaneous fission neutron probability distribution,
ν_{I2} = reduced second factorial moment of the induced fission neutron probability distribution,
ν_{S3} = reduced third factorial moment of the spontaneous fission neutron probability distribution,
ν_{I3} = reduced third factorial moment of the induced fission neutron probability distribution,
λ = the neutron detector decay constant for fission neutrons,
Ω = the inspection time interval width in μs, and

WW2 = the reduction in the number of "double" neutrons observed due to the finite inspection interval, Ω, and the detector die-away time constant, λ.
WW3 = the reduction in the number of "triple" neutrons observed due to the finite inspection interval, Ω, and the detector die-away time constant, λ.

For the active interrogation of ^{235}U samples using delayed neutrons we make the following assumptions:

1. The spontaneous fission rate for ^{235}U is zero, i.e. Fs=0.

2. The Feynman observables are due solely to delayed neutrons and neutron induced fission of the ^{235}U fissile material.

3. The random neutron source is the delayed neutrons from fission products, and we replace $S\alpha$ with S_d.

Under the above assumptions, the above formalism simplifies to describe delayed neutron interrogation measurements as follows:

$$R1 = \varepsilon * M_L * S_d,$$

$$R2 = (\varepsilon^2) *[M_L{}^2*\{(M_L -1)*\nu_{I2} /(\nu_{I1} -1)\}* S_d] \text{ and,}$$

$$R3 = (\varepsilon^3) * M_L{}^3*\{(M_L-1) /(\nu_{I1} -1) *[\nu_{I3}+2*(M_L-1) *\nu_{I2}{}^2/(\nu_{I1} -1) \}* S_d].$$

The ratios N2/N1 and N3/N1 eliminate the delayed neutron rate S_d and remain functions of ε and M_L:

$$R2/R1 = (\varepsilon)*[M_L *\{(M_L -1) *\nu_{I2} /(\nu_{I1} -1)\}] \text{ and,}$$

$$R3/R1 = (\varepsilon^2)* M_L{}^2*\{(M_L -1) /(\nu_{I1} -1) *[\nu_{I3}+2*(M_L -1) *\nu_{I2}{}^2/(\nu_{I1} -1)\}]\}.$$

8. Comparison of N2/N1 and N3/N1 to the Hage-Cifarelli Model.

The ratios N2/N1 and N3/N1 are dependent upon only ε and M_L within the Hage-Cifarelli model. For the measurements reported here, we estimate the fission neutron detection probability of the neutron detector to be ε = 0.27 from N2/N1 and N3/N1 ratios measured with a ^{252}Cf neutron source. The reduced moments of the neutron emission probabilities were derived from the thermal neutron emission probabilities of Bolden and Hines [10] for ^{235}U according to the method of Terrel[11]. The thermal neutron emission probabilities were first fit with a gausian function. Then the centroid of the distribution was shifted by the difference between the average number of neutrons from thermal neutron induced fission (ν_{I1} = 2.41) to that for fission energy neutron induced fission (ν_{I1} = 2.62), and the gausian distribution calculated with all other parameters obtained from the fit to the thermal distribution. The reduced factorial moments of the distribution were then formed and are listed in Table II.

Table II. The first, second and third factorial moments of the neutron probability distribution .

Neutron probability moment	Value
ν_{I1}	2.62
ν_{I2}	2.76
ν_{I3}	1.52

The results for N2/N1 and N3/N1 for the delayed neutron interrogation measurements are shown in Fig. 7, with the solid line representing the calculation for $\varepsilon = 0.27$. The values for

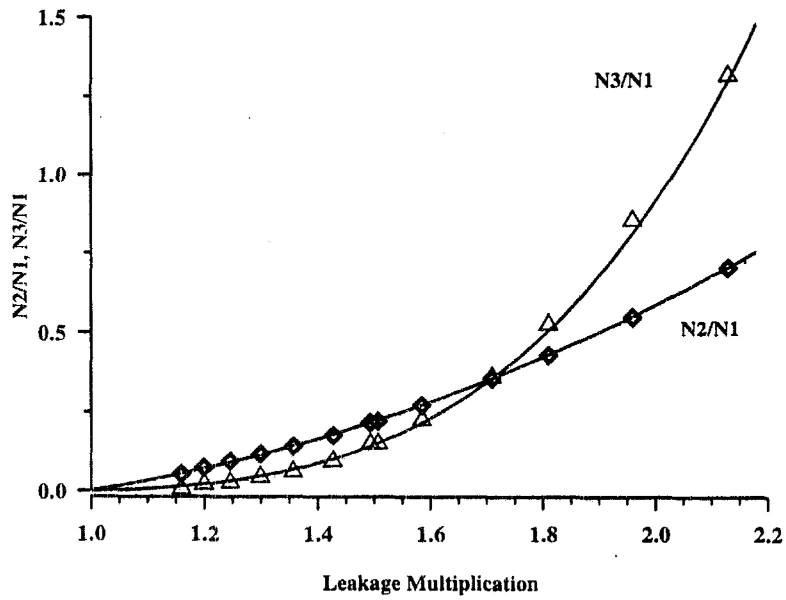


Fig. 7. The variation of the ratio of the Feynman Variance observables N2/N1 (diamonds) and N3 /N1 (triangles) obtained with delayed neutron interrogation with the leakage multiplication obtained from the calculations from the Hage-Cifarelli model.

multiplication were adjusted to optimize simultaneously the agreement for both N2/N1 and N3/N1 ratios. The values for the multiplication are listed in Table III under Multiplication

Table III. The values for the experimental leakage multiplications, experimental total multiplications and MCNP K-Code multiplications for the twelve configurations with indicated mass and radii.

Mass (gm)	Radius (cm)	Multiplication Leakage	Multiplication Total	Multiplication One-Dant
592.0	2.00	1.16	1.26	1.29
932.7	2.34	1.21	1.34	1.36
1382.6	2.67	1.26	1.42	1.43
1961.9	3.00	1.31	1.50	1.52
2683.4	3.34	1.36	1.59	1.61
3570.8	3.67	1.42	1.69	1.73
4625.4	4.00	1.50	1.82	1.85
5868.3	4.34	1.59	1.96	1.99
7327.2	4.67	1.71	2.17	2.15
9020.0	5.00	1.81	2.36	2.35
10920.0	5.34	1.96	2.58	2.57
13722.0	5.67	2.13	2.86	2.84

Leakage. The multiplication reported above is known as the "leakage multiplication" (M_L) or "net multiplication"[12], that is the net gain in neutrons per source neutrons. According to Serber [12] the total multiplication (M_T), the total number of neutrons in the system per initial neutron, is related to the leakage multiplication by:

$$M_T = (M_L * \nu - 1 - \alpha)/(\nu - 1 - \alpha),$$

where ν is the average number of neutrons emitted /fission and α is the ratio of neutron capture cross section to fission cross section. For the case of ^{235}U, we assume α =0.03, and ν =2.62. Listed in Table III are the values for the total multiplication obtained with the above relation, along with total multiplications calculated with the One-Dant Code for the twelve HEU metal configurations. The total multiplication values for the twelve HEU samples from delayed neutron interrogation and One-Dant Code results are displayed in Fig. 8. A

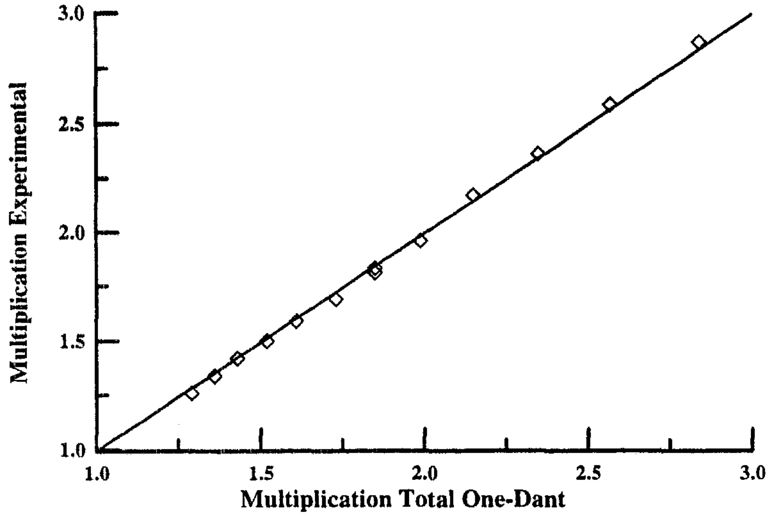
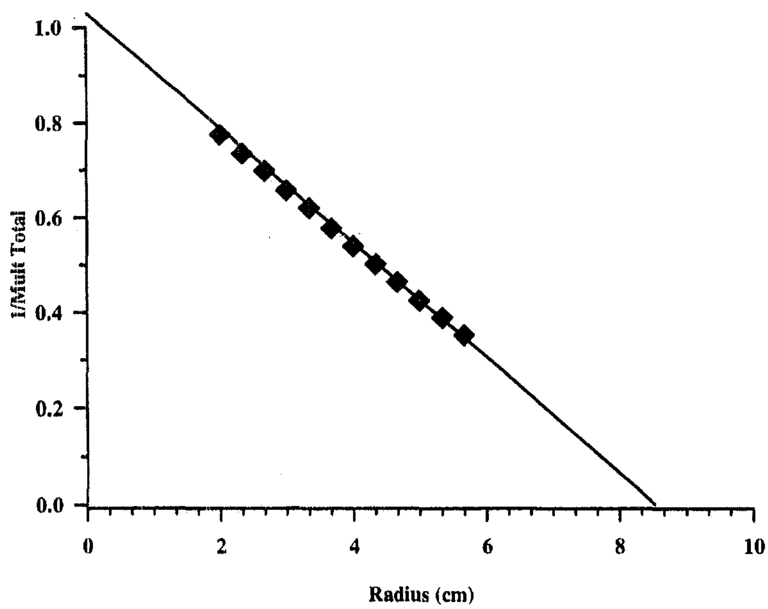


Fig. 8. The multiplication values for the twelve configurations of HEU spheres extracted from the delayed neutron interrogation compared to One Dant calculations. The solid line represents a linear fit to the data.

least squares fit to the data yields the following relationship:

$$M_T = 0.995 * M_{T\ \text{One-Dant}} .$$

In Fig. 9 is shown the relationship between the experimentally determined total multiplication



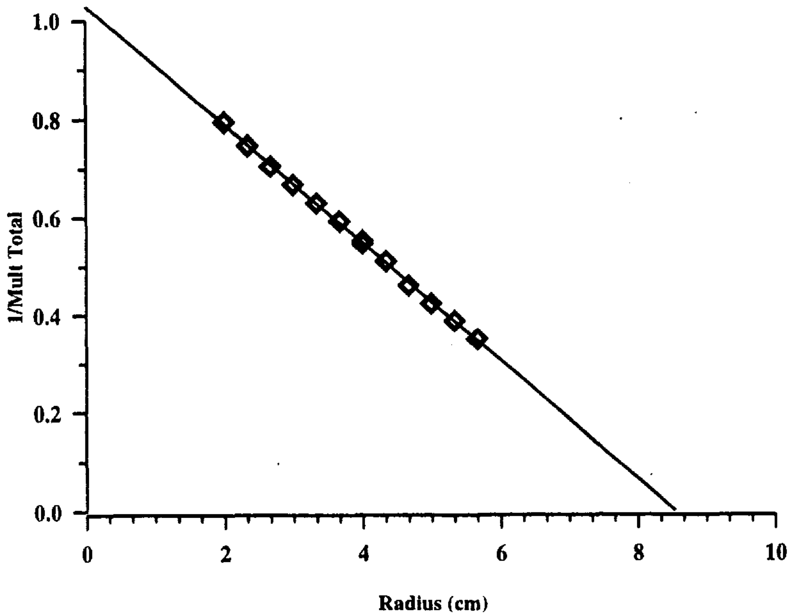


Fig. 9. The values of $1/M_T$ vs the radius of the HEU system.

and the radii of the HEU samples. The solid line represents a least squares fit to the data of the function

$$y = 1.031 - 0.121x.$$

Evaluating this function at y = 0, i.e. $1/M_T = 0$, yields the radius at which the associated HEU mass has reached the "critical mass". The value of radius of 8.545 ± 0.075 cm is obtained, which corresponds to a value of the critical mass of 48.98 ± 0.43 kg. The critical mass for a 93.71% enriched HEU sphere is 49.12 ± 0.15 according to the compilation of Paxton and Pruvost[13].

9. Mass Determination

According to the Hage-Cifarelli model, the singles counting rate N1/sec should vary as

$$N1/sec = \varepsilon * M_L * S_d,$$

where S_d is the number of delayed neutrons emitted per second, M_L is the leakage multiplication and ε is the neutron detection probability. S_d should be proportional to the 14-MeV neutron flux, the 14-MeV n-fission cross-section, the number of delayed neutrons emitted per fission, the mass of the ^{235}U and the multiplication. Thus the following relationship is obtained:

$$N1/sec = M_L*(Mass*M_L*A), \text{ or}$$

$$= A*M_L^2*Mass,$$

where A (a constant) includes ε, the neutron flux, the n-fission cross section and the number of delayed neutrons per fission. The variation of N1/sec divided by the square of the multiplication, as a function of mass, is shown in Fig. 9, along with a linear least squares fit to the data.

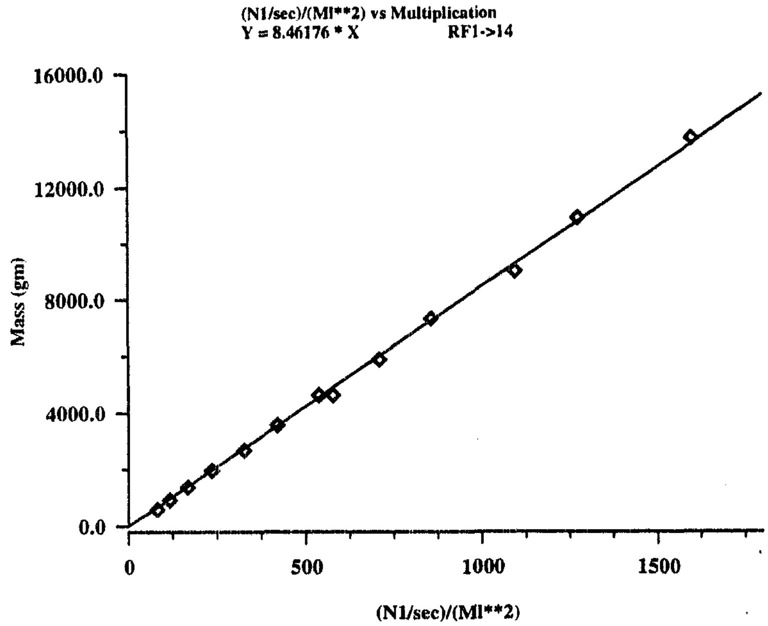


Fig. 9. N1/sec values divided by the multiplication squared vs. mass of the twelve HEU configurations for delayed neutron interrogation.

Solving for the mass yields the following:

$$\text{Mass} = 8.462*(\text{N1/sec})/(M_L^2).$$

This simple linear relationship between the mass and (N1/sec)/(M_L^2) is indicative that the fission products, and hence the source of delayed neutrons, are evenly distributed throughout the volume of the HEU samples.

10. Summary

A new method to determine neutron multiplication values using delayed neutrons has been developed and applied to twelve bare HEU metal systems of solid spheres. These multiplication values are found to be closely related to those obtained from the One-Dant computer code. This method of measurement and analysis places the determination of multiplication of HEU systems on an equal basis with plutonium systems driven by spontaneously fissioning ^{240}Pu.

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